Eastern Asian emissions of anthropogenic halocarbons deduced from aircraft concentration data

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Short title: EASTERN ASIAN HALOCARBON EMISSIONS

Abstract. Halocarbons are a class of ozone-depleting gases whose production is banned in developed countries by the Montreal Protocol, and is subject to a series of restrictions in developing countries. Recent aircraft measurements of Asian outflow over the western Pacific (March-April, 2001) offer important constraints for estimating emissions from eastern Asia (China, Japan, and Korea), a region whose consumption of halocarbons is particularly uncertain. We report the first aircraft-deduced emission estimates of selected halocarbons from China, Japan, and Korea. We calculate an eastern Asian carbon tetrachloride (CCl_4) source of 18.5 Gg yr^{-1}, considerably larger than previous estimates and a major contributor to the global budget for this gas. Our results for methyl chloroform (CH_3CCl_3) are in general agreement with inventories for 1999 derived from government records, and our emission estimates for CFC-11 and CFC-12 are consistent with those calculated from records of production and consumption. For Halon 1211 we find only a strong local source originating from the Shanghai area. Our emissions result in a 40% increase in ozone depletion potential (ODP) from previous estimates of eastern Asia for the gases studied, amounting to a $\simeq$10% global increase in ODP.

Introduction

The role of halocarbons in the destruction of stratospheric ozone is well established [*WMO/UNEP*, 1999]. The 1987 Montreal Protocol outlined approaches to ultimately cease the production of these compounds. The long chemical lifetime of halocarbons allows them to travel from the surface to the stratosphere where they are subsequently photolysed, releasing halogens that catalyse ozone-depleting chemical reactions. The relative strength of a halocarbon gas to destroy stratospheric ozone is given by the ozone depleting potential (ODP), defined as the ratio of the net chemical destruction of ozone by a specified mass of that gas to a similar mass of CFC-11 (Table 1).

To estimate emissions of halocarbons from eastern Asia we use measurements of halocarbons [*Blake et al.*, 1996] and CO [*Sachse et al.*, 1987] from the NASA TRACE-P Global Troposphere Experiment [*Jacob et al.*, 2003] (Figure 1), a two-aircraft mission flown over the western Pacific during Spring 2001. The aircraft operated out of Hong Kong and Japan, and flights covered the Pacific rim between 12 and 45°N. These data provide considerable geographical coverage of Asian outflow, and therefore are more likely to characterise regional emissions of halocarbons from eastern Asia than measurements at a single site. Boundary layer flights (0−2 km) west of 150°E sampled fresh continental outflow. We utilize observed relationships between particular halocarbons and CO to determine emissions of the halocarbons. Sources of CO from fuel consumption (fossil fuel and biofuel) are relatively well known and are in general collocated with the halocarbon sources. Loss of CO is by oxidation by OH, resulting in an atmospheric lifetime of a few months. Provided there is a strong correlation between CO

and a particular halocarbon, one can infer the halocarbon emission from the halocarbon:CO relationship. We use this methodology to quantify emissions of halocarbons from eastern Asia.

Methyl Chloroform (CH_3CCl_3)

Methyl chloroform (ODP=0.1) has a relatively minor role in stratospheric ozone depletion but it is most important as the standard proxy for the global concentration of the hydroxyl radical (OH), the main atmospheric oxidant for many environmentally important gases. Measurements of CH_3CCl_3 concentration have been used to infer trends in the oxidizing power of the atmosphere [*Spivakovsky et al.*, 1990; *Spivakovsky et al.*, 2000, *Krol et al.*, 1998, *Montzka et al.*, 2000, *Prinn et al.*, 2001].

The Montreal Protocol called for a total ban on the production and sales of CH_3CCl_3 in developed countries by 1996. Beginning in 2003, the first in a series of restrictions on consumption in developing countries takes effect. Prior to the Montreal Protocol, accurate emission estimates of CH_3CCl_3 were calculated from manufacturers' records of production and sales [*Midgley and McCulloch*, 1995; *McCulloch and Midgley*, 2001]. Now, however, emissions from developed countries are generally thought to represent slow release from legal stockpiles accumulated prior to the ban [*Barnes et al.*, 2003; *Krol et al.*, 2003], and are consequently difficult to quantify. Recent studies have attempted to determine emissions of CH_3CCl_3 from Europe and North America using fixed long-term data sets [*Barnes et al.*, 2003] or from aircraft data during short intensive measurement campaigns [*Krol et al.*, 2003]. These studies find a significant source of CH_3CCl_3 from Europe and North America, casting serious doubt on studies of trends in the oxidising capacity of the atmosphere that rely on government reports of zero

emissions in these regions [*Prinn et al.*, 2001].

We use boundary layer (0−2 km) aircraft observations taken directly downwind of the Asian continent during the TRACE-P mission, and find that concentrations of CH_3CCl_3 and CO are correlated in the Asian outflow (Figure 1A), suggesting that these two gases originate from common areas. Outflow of CO below 2 km during TRACE-P was mainly anthropogenic, with minimal contribution from biomass burning [*Liu et al.*, 2002]. Analysis of five-day isentropic back-trajectories [*Fuelberg et al.*, 2002] for the largest 5% ($\geq$45.9 pptv) of CH_3CCl_3 concentrations shows that they originate largely from South Korea and the area around Shanghai (Figure 2A).

CH_3CCl_3 and CO both have background latitudinal gradients, reflecting their sources at northern mid-latitudes and the higher OH concentrations in the tropics. The influence of these gradients on the CH_3CCl_3:CO relationship is removed by subtracting background values (defined as the 20th percentile) for 5° latitude bins. Measurements with this background removed are denoted by Δ. We further remove statistical outliers ($\geq$95th percentile of CH_3CCl_3 and CO) so that our derived emissions are not influenced by local sources and are more representative of the region. There is a statistically significant relationship between $\Delta CH_3CCl_3$and ΔCO using all the data (Figure 3A), validating the use of a CO inventory to determine emissions of CH_3CCl_3.

We use a CO emission inventory for eastern Asia during TRACE-P that is based on detailed a priori mission-specific inventories [*Heald et al.*, 2002; *Streets et al.*, 2002] and has been refined with a formal inverse model analysis constrained by concentration data [*Palmer et al.*, 2003]. *A posteriori* anthropogenic emissions from China (142.3$\pm$6.0 Tg CO yr^{-1}) are

approximately 30% greater than *a priori* values [*Palmer et al.*, 2003]. *A posteriori* emissions for Korea and Japan are not significantly higher than the a priori so that the *a priori* estimates are retained here (9.3±1.6 Tg CO yr^{-1} for Korea and 8.5±3.5 Tg CO yr^{-1} for Japan) [*Streets et al.*, 2002]. Combining information about CH_3CCl_3:CO calculated using all data (Figure 3A) with the CO emission estimate for eastern Asia leads to a CH_3CCl_3 emission estimate for eastern Asia of 12.2±0.9 Gg yr^{-1}.

We further refine this emission estimate for eastern Asia by disaggregating contributions from China (CH), Japan (JP), and Korea (KR), using five-day isentropic back-trajectories to classify the origin of sampled air masses (Figure 2A). There are instances when air passes over more than one country, which we take into account by including the additional classifications of CHKR, CHJP, and KRJP, and instances when air originates from the marine boundary layer (10% of all back-trajectories) with values typical of the global background. We consider only observations whose back-trajectories pass over countries at altitudes below 850 hPa. Most air masses (44%) originate from mainland China. Combining the CH_3CCl_3:CO slope for China (0.013 pptv/ppbv; Figure 3) with the CO emission estimate for China leads to CH_3CCl_3 emissions of 8.8±0.8 Gg yr^{-1} for that country. The CH_3CCl_3:CO slopes for air masses originating from Japan (0.023 pptv/ppbv) and Korea (0.044 pptv/ppbv) are much higher than for China. From these slopes we estimate CH_3CCl_3 emissions of 0.9±0.5 Gg yr^{-1} for Japan and 2.0±0.6 Gg yr^{-1} for Korea. The CH_3CCl_3:CO slopes for air masses that pass over more than one country (Figure 3) are intermediate and consistent with the values derived above. However, the relatively weak CH_3CCl_3:CO correlation for air masses originating from Japan means that our corresponding emission estimate is highly uncertain.

Our emission estimate for eastern Asia in 2001 is in good agreement with emission estimates for the 'Far East' (11 Gg yr^{-1}) [*McCulloch and Midgley*, 2001] that have been extrapolated from government data for previous years, and with consumption data from the United Nations (12.5 Gg yr^{-1}) [*UNEP*, 2002], derived from government records. Our CH_3CCl_3 emission estimates for particular countries within eastern Asia do not agree with previous emission estimates. Past studies have assumed zero emissions from Japan in recent years, with emissions from eastern Asia due largely to China and Korea [*McCulloch and Midgley*, 2001]. Records of consumption have been generally thought to provide the most accurate representation of emissions. The value we report for China during 2001 is $\simeq$50% larger than the value for consumption reported by the UN for 1999 (records of consumption for later years are incomplete), suggesting that either the government records of consumption are not a good proxy for emissions, or a steep increase in emissions. The UN estimate for Korean emissions of CH_3CCl_3 is 6 Gg yr^{-1}.

Carbon Tetrachloride (CCl_4)

Emissions of CCl_4 (ODP=1.1) originate primarily from its use as a chemical feedstock for the production of CFC-11. The Montreal Protocol called for a total ban of this gas in developed countries by 1996, while restrictions in developing countries are scheduled to start in 2005. The total Asian source of CCl_4 in 1999 according to the UN is 17 Gg yr^{-1} [*UNEP*, 2002], with India contributing more than 80% to this value. Using the method outlined above, we calculate emissions from eastern Asia of 18.5$\pm$1.4 Gg yr^{-1}. Because our CCl_4:CO relationship is statistically significant (Table 1) and our five-day back-trajectories do not pass over India (not

shown), we can exclude the possibility that Indian emissions of CCl_4 influence the calculated eastern Asian signal. The large discrepancy between our result and previous estimates of CCl_4 emissions from eastern Asia highlights serious shortcomings in our knowledge of the sources of this gas.

Our method used to classify back-trajectories was successful in determining a unique statistically significant slope for China (Table 1), corresponding to CCl_4 emissions of 14.9 ± 1.4 Gg yr^{-1} and representing 80% of the total eastern Asian emission of this gas. The UN report emissions of 0.1 Gg yr^{-1} for China. There are also unique slopes for Korea and Japan, with CHKR more representative of China, and with KRJP and CHJP more representative of the Japanese slope (not shown). Emissions of CCl_4 from Korea and Japan are 2.6 ± 0.8 Gg yr^{-1} and 1.2 ± 0.6 Gg yr^{-1}, respectively. The UN report Korean emissions of 1.3 Gg yr^{-1} and Japanese emissions of 0.1 Gg yr^{-1}.

Halon 1211

Halons are amongst the most effective gases in destroying stratospheric ozone, with ODPs $\gg 1$. Emissions of halons from developed countries were banned in 1994, while emissions from developing countries are currently frozen at their values for 1995-1997. Three halons were measured during TRACE-P: Halon 1211, halon 1301, and halon 2402. Halon 1211 was the only halon that was correlated with CO. Halon 1211 (CF_2BrCl, ODP=3.0) is used as a fire retardant, and China is one of the few countries in the world that still produce this compound. Analysis of back-trajectories for the largest 5% ($\geq$6.1 pptv) of Halon 1211 concentrations shows they originate from central China, and in particular a large cluster of trajectories originate from the

area around Shanghai (Figure 2B). We find that by removing the highest 5% of concentration data, in order to obtain a Halon 1211:CO slope more representative of the regional signal, we effectively remove any useful signal that is significantly different from the values typical of the background atmosphere (0.003 pptv/ppbv). The slope for the top 5% of concentration data (n=88, r=0.62) is 0.023 pptv/ppbv, but we lack information on the Shanghai source of CO that would be needed to infer halon 1211 emissions.

Chloroflurocarbons

Traditional uses for CFCs include air-conditioning, refrigeration, and foam blowing. The Montreal Protocol imposed a total ban on production and consumption of these gases in developed countries in 1996; developing countries are subject to a series of restrictions, beginning in 2003 with a 20% reduction in production and consumption relative to 1998-2000 values. Four CFCs were measured during TRACE-P. We find statistically significant correlations with CO for CFC-11 ($CFCl_3$, ODP = 1.0) and CFC-12 (CF_2Cl_2, ODP = 1.0) (Table 1).

Eastern Asian emissions of CFC-11 and CFC-12, using the slopes shown in Table 1, are estimated to be 25.9$\pm$2.2 Gg yr^{-1} and 33.9$\pm$2.9 Gg yr^{-1}, respectively. For these gases, the CFC:CO relationship for individual countries is better correlated than the slope for eastern Asia (Table 1), suggesting highly heterogeneous sources among the different countries. Chinese, Korean, and Japanese sources are 18.9$\pm$1.9, 3.20$\pm$0.70, and 3.2$\pm$1.0 Gg yr^{-1}, respectively for CFC-11; and 24.0$\pm$2.5, 4.0$\pm$1.4, and 3.0$\pm$1.5 Gg yr^{-1}, respectively for CFC-12. Eastern Asian emissions of CFC-11 calculated from marketing records for 2000 are 17.9 Gg yr^{-1} [*McCulloch et al.*, 2001], representing 24% of the estimated global source of this gas; contributing emissions

from China, Korea, and Japan are 7.2, 2.6, and 8.1 Gg yr^{-1}, respectively. Total emissions for eastern Asia and national emissions for Korea are in good agreement with our results, however, the estimated emissions for China are less than half those we report, while the estimated emissions for Japan are more than double those we report. Eastern Asian emissions of CFC-12 derived from sales and production records for 2000 are 37.2 Gg yr^{-1} [*McCulloch et al.*, 2003], representing 28% of the estimated global source for this gas; emissions from China, Korea, and Japan are 20.3, 5.6, and 11.4 Gg yr^{-1}. These results agree remarkably well with the estimates we report, given the uncertainties of both methods. We are less confident of our estimate for Japanese emissions due to the relatively small correlation with CO and the relatively few observations determining the CFC-12:CO slope.

Implications of Results

Determining long-term trends in the OH radical from the CH_3CCl_3 proxy requires accurate knowledge of CH_3CCl_3 emissions. Eastern Asia is one region where knowledge of emissions of CH_3CCl_3 is highly uncertain. Our emissions for eastern Asia are consistent with those assumed in a recent study of the decadal OH trend [*Prinn et al.*, 2001]. Recent work has shown significant ongoing emissions of CH_3CCl_3 from Europe [*Krol et al.*, 2003; *Barnes et al.*, 2003] and North America [*Barnes et al.*, 2003], with a combined total twice the magnitude of our emissions from eastern Asia. European emissions [*Krol et al.*, 2003] were estimated with an *ad hoc* method using the highest observed concentrations of CH_3CCl_3 and thus likely to represent an upper limit. Our confirmation that eastern Asian emissions of CH_3CCl_3 assumed in recent studies are approximately correct is consistent with the idea that OH has no significant trend

over the past decade.

We estimate a total annual halocarbon source (CH_3CCl_3, CCl_4, CFC-11, CFC-12) from eastern Asia of 81.6 ODP Gg yr^{-1}, a 40% increase in ODP from previous estimates [*McCulloch and Midgley*, 2001; *McCulloch et al.*, 2001; *McCulloch et al.*, 2003; *Simmonds et al.*, 1998] for these gases. This regional increase amounts to a $\simeq$10% increase in global ODP for these gases, using published global emission estimates [*McCulloch and Midgley*, 2001; *McCulloch et al.*, 2001; *McCulloch et al.*, 2003; *Simmonds et al.*, 1998]. This global increase in ODP is likely a conservative estimate, owing to our use of estimated emission of CCl_4 from 1995 [*Simmonds et al.*, 1998] in the absence of more recent data (long-term trend of concentration measurements suggests that present-day emissions are lower than 1995 emissions [*Montzka et al.*, 1999]). In 1995, the halocarbons studied here are estimated to account for $\simeq$70% of the total global ODP [*WMO/UNEP*, 1999].

More generally, the method we have used to construct regional-scale emission inventories of anthropogenic halocarbons from aircraft concentration data in regional outflow does not rely on government records, and therefore can be used as an independent test of reported values for production or consumption. This method also has considerable potential to monitor the magnitude and trends of emissions of a wide range of environmentally important gases.

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Figure Captions

Figure 1 Geographical distributions of **A** CH_3CCl_3, **B** CCl_4, **C** Halon 1211, **D** CFC-11, **E** CFC-12, and **F** CO concentrations measured in the boundary layer ($0-2$ km) during the TRACE-P aircraft campaign (March-April 2001).

Figure 2 Five-day isentropic back-trajectories indicate the origin of the highest five percent of **A** CH_3CCl_3 and **B** Halon 1211 concentrations measured in the boundary layer ($0-2$ km) during the TRACE-P aircraft campaign (March-April 2001). Blue squares represent the measurement locations and open circles along the back-trajectories denote 12-hour increments.

Figure 3 Reduced Major Axis Regression (RMAR) [*Hirsch and Gilroy*, 1984] of CH_3CCl_3 and CO enhancements over their background values (ΔCH_3CCl_3 and ΔCO). **A** All available data. Green circles denote data $\geq$95th percentile of CH_3CCl_3 and CO concentrations, and are removed from the RMAR calculations. **B** Air masses sampled downwind of China (CH); **C** air masses sampled downwind of Korea (KR), and downwind of both China and Korea (CHKR); and **D** air masses sampled downwind of Japan (JP), downwind of both China and Japan (CHJP), and downwind of both Korea and Japan (KRJP). Air masses whose five-day back-trajectories do not coincide with any country or pass over a country above an altitude greater than 850 hPa are not considered here.

Table Captions

Table 1 Halocarbon:CO relationships measured in Asian outflow during TRACE-P. Reduced Major Axis Regression (RMAR) slopes were computed after first subtracting the latitudinally dependent background (defined as the 20th percentile of the observations) from both the halocarbon and CO, and considering only data $<$95th percentile. Asterisks denote a halocarbon:CO relationship that does not differ from that of the background atmosphere.

Figure 1

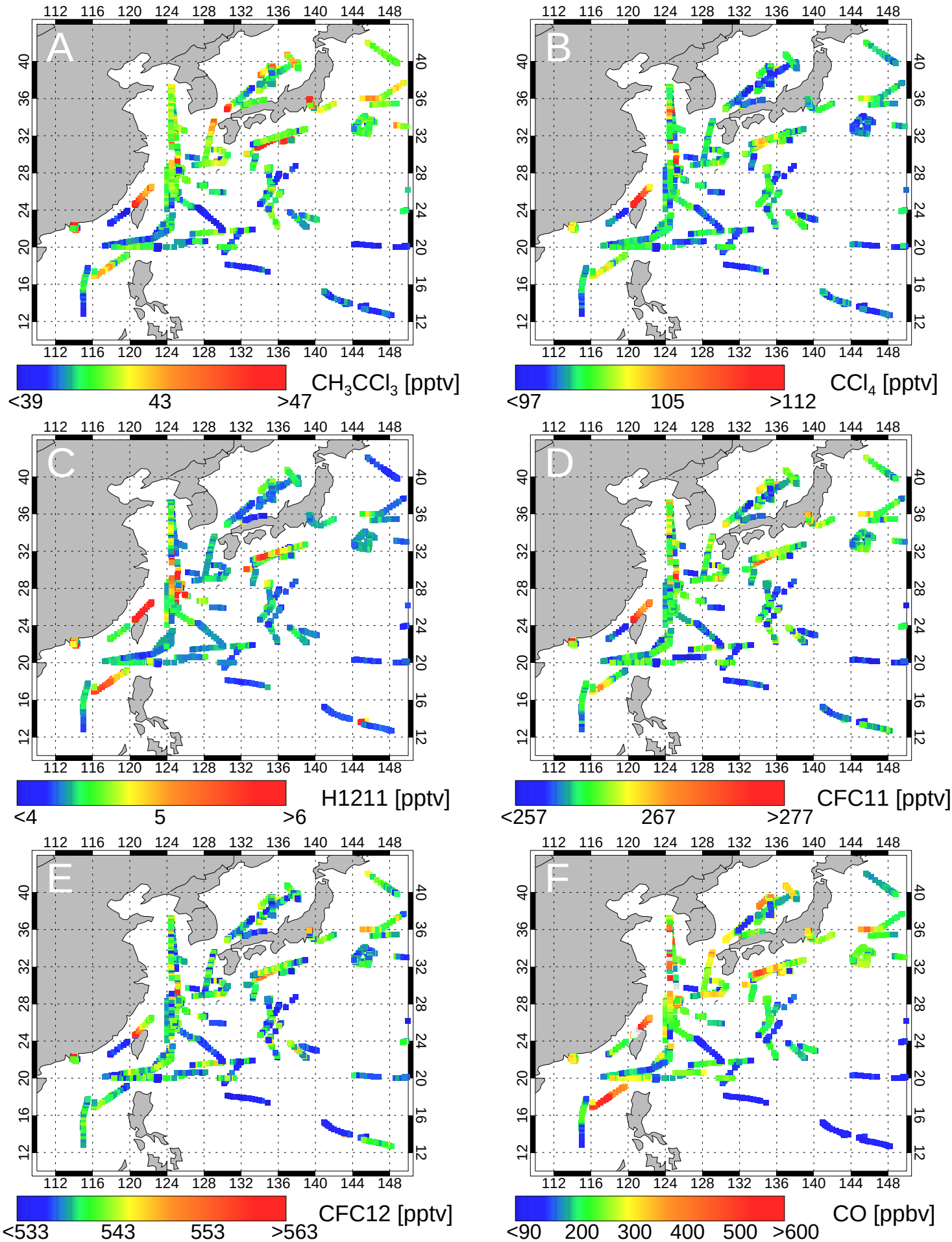

A
B
C
D
E
F
112 116 120 124 128 132 136 140 144 148
40 36 32 28 24 20 16 12
CH_3CCl_3 [pptv]
<39 43 >47
CCl_4 [pptv]
<97 105 >112
H1211 [pptv]
<4 5 >6
CFC11 [pptv]
<257 267 >277
CFC12 [pptv]
<533 543 553 >563
CO [ppbv]
<90 200 300 400 500 >600

Figure 2

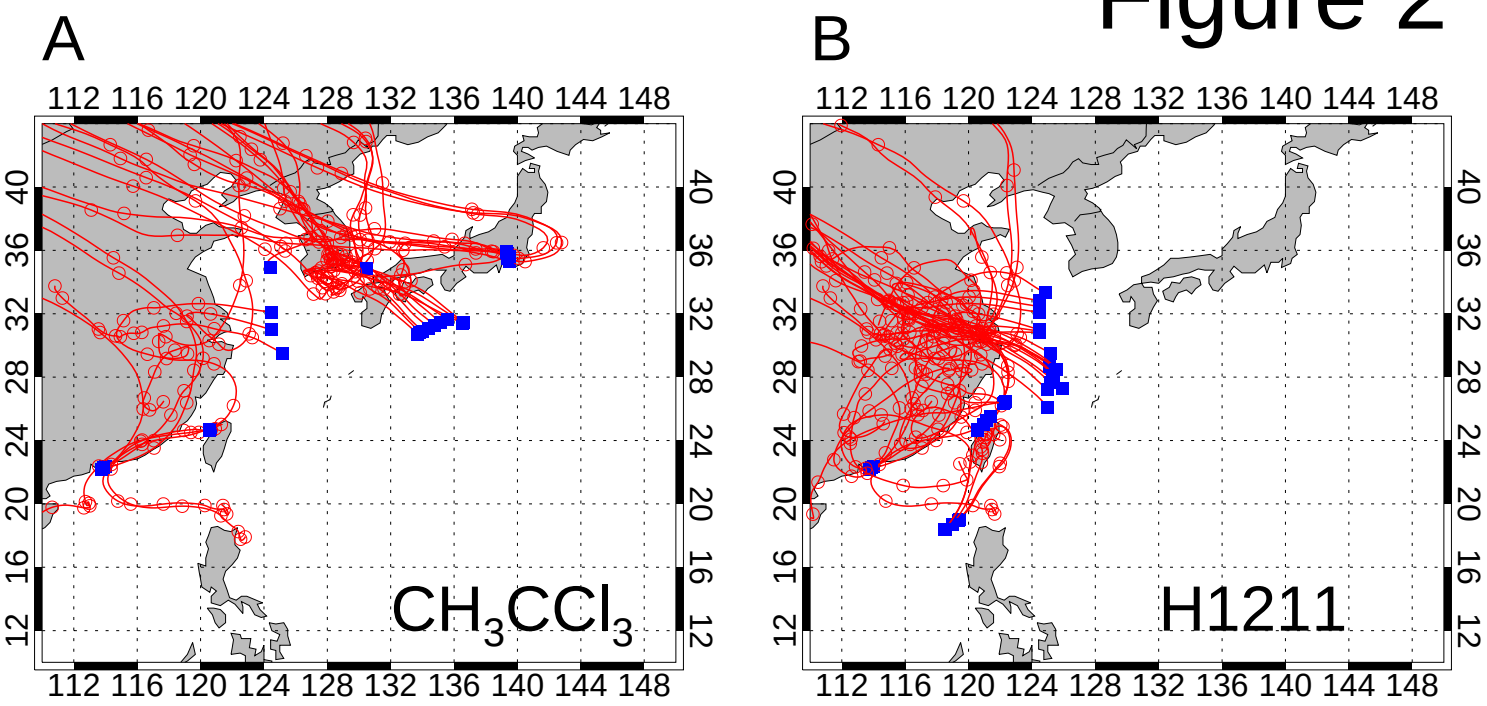
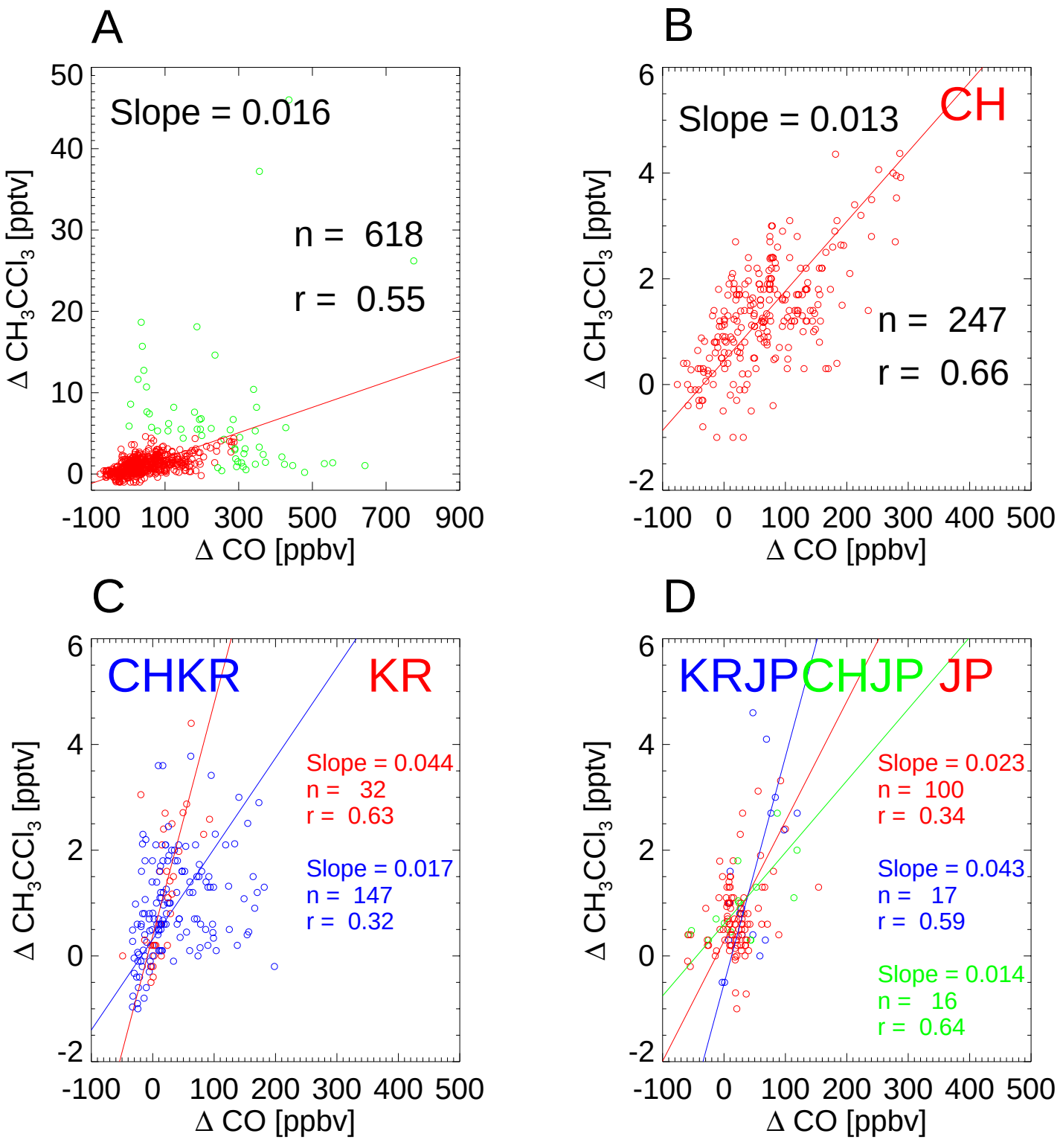

A
B
112 116 120 124 128 132 136 140 144 148
40 36 32 28 24 20 16 12
CH_3CCl_3
H1211

Figure 3


A
Slope = 0.016
n = 618
r = 0.55
Δ CH3CCl3 [pptv]
Δ CO [ppbv]
B
Slope = 0.013
CH
n = 247
r = 0.66
Δ CH3CCl3 [pptv]
Δ CO [ppbv]
C
CHKR
KR
Slope = 0.044
n = 32
r = 0.63
Slope = 0.017
n = 147
r = 0.32
Δ CH3CCl3 [pptv]
Δ CO [ppbv]
D
KRJP
CHJP
JP
Slope = 0.023
n = 100
r = 0.34
Slope = 0.043
n = 17
r = 0.59
Slope = 0.014
n = 16
r = 0.64
Δ CH3CCl3 [pptv]
Δ CO [ppbv]

	ODP	Eastern Asia			China			Korea			Japan		
		Number of data n	RMAR 10^{-3} [pptv/ppbv]	Correlation r	n	RMAR	r	n	RMAR	r	n	RMAR	r
CH_3CCl_3	0.1	618	16±0.48	0.55	247	13±0.65	0.66	32	44±6.2	0.63	100	23±2.1	0.34
CCl_4	1.1	620	21±0.63	0.60	239	19±0.95	0.64	36	50±6.5	0.59	101	25±2.3	0.47
Halon 1211 (CF_2BrCl)	3.0	556	3±0.15	0.54	*	*	*	*	*	*	*	*	*
CFC11 ($CFCl_3$)	1.0	611	33±1.7	0.46	238	27±1.62	0.46	34	69±9.7	0.56	98	50±4.0	0.56
CFC12 (CF_2Cl_2)	1.0	613	49±2,5	0.38	246	39±2.34	0.43	33	99±16.8	0.56	97	83±7.5	0.44